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# THE ROLE OF STABLE ISOTOPES IN GEOCHEMISTRIES OF ALL KINDS

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Although I was surprised by the invitation to write a prefatory chapter for the *Annual Review of Earth and Planetary Sciences*, I decided to accept this opportunity to write about my experiences as a participant in the isotope geochemistry revolution that occurred 40–50 years ago. My own experiences during the early stages of my life were uniquely intertwined with those of many of the leaders of this revolution, and thus I was allowed to witness and participate in many remarkable changes in the science as a whole. In addition, I thought that my contribution might be of some interest to the members of a younger generation, to help them judge whether a scientific career could be interesting, rewarding, and worth working hard for to accomplish something. This chapter also gives me the opportunity to express my appreciation to some of the teachers, early colleagues, and students whose influences were important in my life.

I was born in 1919 in a village close to Kobryn, which is now part of Belarus. At that time, the question of who owned Kobryn was still causing considerable armed conflict. Prior to World War I, it had been part of the Russian Empire. After the war, it became part of Poland. Being a resident of that part of the world during those times was not pleasant, nor did the future appear to be all that promising. Consequently, my father, who had lived in New York for a few years as a teenager and fully understood the advantages of living in the western world, decided to leave Poland. Because US immigration laws in those days included a strict quota system requiring a long wait, my father and mother decided to go to Canada. My father left Kobryn in 1926, and in 1927 my mother, my two older sisters, and I joined him. My gentle and very caring mother was already very ill at that time as a consequence of the horrors she and my father and sisters suffered during World War I. She passed away in 1928. This tragic

loss was somehow made easier by the love and care that my older sisters and my father gave me during the next important and formative years of my life. It goes without saying that the decision my parents made to immigrate to Canada saved us from certain destruction during World War II, a fate that overcame the rest of our family who remained in Kobryn.

The 1930s were plagued by the Great Depression, and career opportunities were not readily available to newly arrived immigrants who came without a special skill or fluency in the English language. Although my father found work and earned enough to feed his family, I understood quite early in life that if I wished to continue my studies beyond high school, I would have to earn some money to pay for this privilege myself.

During my high school days, I managed to get a paper route for the Winnipeg Free Press and did a good job at it. I also managed during the years that followed to find temporary jobs, ranging from working on the Boardwalk on Winnipeg Beach to spraying pesticides for the war against the mosquitoes that annually plagued Winnipeg well into each summer. I appreciated these opportunities to be useful and earn money at the same time. I also knew they offered me a way to continue my education if I decided to attend the university. Money problems were the only thing that could have prevented it.

Since my earliest schooling, my favorite subjects were mathematics and what at that time passed for popular science and engineering. As a youngster, engineering was much more understandable to me than were other subjects, and I was inclined toward a career in it. Realistically, the uncertainties in my future never allowed me to get too set in my hopes for my life. From early on, many more-immediate family problems were there to contend with, and I was not willing to exclude any possibility.

Throughout this period, however, my family made sure that whatever funds I earned would be put away for my education. I also made sure that the courses I took in high school would meet the entrance requirements for the University of Manitoba.

Generally, I enjoyed my high school years, and my favorite subjects continued to be related to the sciences. I was fortunate to have a physics teacher who did a minimum of formal lecturing, and who depended on our learning physics by working problem sets, which was actually a very good idea.

The University of Manitoba offered two degrees in sciences: a regular four-year Bachelor of Science degree and a five-year Honors Bachelor of Science degree. For the regular degree you selected four optional subjects. For the honors degree, you were required during your last two years to specialize in two subjects. I selected math, physics, chemistry, and geology for my optional subjects, and chemistry and geology for my honors degree requirements.

There were just three of us in the geology honors degree program. Strangely, I was able to qualify for this degree with very little field geology experience.

Most of the field experience many of the geology majors had was gained during summer months by working for the Manitoba Geological Survey. Somehow I missed the financial rewards and the important field experiences that these summer jobs offered. During my final year before graduation, one of the members of the Manitoba Survey gave me the unsolicited advice that I did not have much of a future in geology. This was a devastating blow, and I was very discouraged. However, it turned out to be a blessing in disguise! That summer I visited one of my chemistry professors, Alan N. Campbell, to discuss an examination I had written for his course. I had apparently done well, because our conversation turned to the possibility of whether I might like to do research with him for a Master's degree, the only advanced degree in chemistry available in Manitoba at that time. There were other possible opportunities for someone with a BS degree for gainful employment as a chemist, but I could not resist the opportunity of working with Dr. Campbell. In part because he had no other candidates at that time, he took me on as a graduate student.

At that time, Alan Campbell was particularly interested in comparing the viscosity of molten selenium with that of molten sulfur at a variety of temperatures. He wanted to determine if these two elements, which lie in the same column of the Periodic Table, had similar properties in their molten states. It was then known that molten sulfur underwent a certain degree of polymerization, and the question was whether selenium also polymerizes. To determine the viscosity of the molten selenium, it was necessary to know the densities of the molten selenium at the desired temperatures. While determining the density of molten selenium, we encountered some serious problems with the quality and strength of the pycnometer that we used for this experiment. Every time the high temperature bath inadvertently cooled down, the selenium solidified; reheating the bath and the solid selenium caused enormous stress in the Pyrex pycnometer, causing it to crack. However, hard work and dogged persistence finally paid off, and our objective was accomplished within one year.

The Chemistry faculty deemed this work to be worthy of a Master's degree. Our year of research on this project involved a relatively simple objective, but solving the problem and carrying it through to completion was a thoroughly new and exciting experience for me. I found Dr. Campbell to be a very accessible professor, and we got along quite well together. I did well in my course work and he obviously appreciated the fact that I had worked hard in solving the problem, my first real "hands-on" experience with scientific research. The results of our effort ended up as a small paper in the *Journal of the American Chemical Society*, representing my first bona fide publication. Most importantly, I realized that doing scientific research was not easy, but it was a wonderful way to spend one's time.

Dr. Campbell encouraged me to apply to McGill University to do graduate work toward a PhD. It was gratifying to me that he thought I might have a

future as a chemist, and his encouragement at this critical period of my life was very important. I sent in my application and received an acknowledgment from McGill that I had the proper qualifications for graduate work. I assumed, therefore, that I had been accepted as a graduate student. Early in September of that year, 1942, I dropped by the Canadian Pacific Railroad station in Winnipeg to inquire if there was something I could do to earn my way to Montreal. The man in charge somehow understood that this was important to me, and gave me a job as a "newsy," selling fruit and sandwiches on the train that was to leave that night. I hurriedly packed and arrived at the station that evening, ready to go to Montreal and McGill University.

When I arrived in Montreal I contacted my friend Leo Yaffe, who had also taken his Master's degree with Alan Campbell, and who had been accepted as a graduate student in the Chemistry Department at McGill the previous year. Leo Yaffe eventually carved out a distinguished career for himself as a faculty member at McGill University. He took me in hand and introduced me to his professor, Carl Winkler, in the Chemistry Department. Dr. Winkler had never heard of me, nor was he aware that I had applied to McGill for graduate work. Somehow there was a mix up; I had never been considered for graduate work by the department.

Dr. Winkler obtained my file from the Dean's Office and found that I had written a perfect paper on an advanced physical chemistry exam, a course taken to qualify for my master of science degree. I also had an excellent letter from Dr. Campbell. In addition, Raymond Boyer, a professor of organic chemistry at McGill, seemed very positive about accepting me for graduate school. Dr. Winkler offered to get me a job in Ottawa, but I expressed my preference for staying as a graduate student at McGill. After what seemed to be an inordinate amount of time, they accepted me as a graduate student, but said they couldn't guarantee any financial help. I was willing to gamble, and within a few weeks I was informed that I could help Dr. Boyer in his organic chemistry laboratory course and earn the regular stipend for graduate students. The stipend was sufficient for me to support myself throughout graduate school. Clearly, Dr. Boyer came through for me, in spite of the fact that my past experience would not have qualified me to do a thesis in his field.

One of the major activities in both Dr. Winkler's and Dr. Boyer's laboratories was the study of the formation of RDX, a very important explosive used during World War II. This explosive was produced by what was known as the Bachman process, and their objective was to study its properties and mechanism of formation.

Organic compounds with the complexity of RDX are formed via a number of intermediate steps. The objective of organic chemistry was to isolate and identify these intermediate compounds and determine the mechanism of their

formation. In addition to RDX, the Bachman process produced another, less than desirable final product: a much less-stable explosive called HMX. It was important to determine how to produce a maximum amount of RDX with a minimum amount of HMX.

An effort was made to determine the relative proportions of RDX and HMX produced by utilizing the differences in their rates of destruction by base hydrolysis. However, because the two compounds were intimately intergrown as they came out as solid products of the Bachman reaction, this technique of preferential destruction did not work well.

I don't quite remember how I got involved in this problem of analyzing the relative proportions of RDX and HMX, but I do recall an experience I had in Dr. Campbell's class involving the kinetics of homogeneous reactions, which gave excellent results in determining the order of the reaction. I decided to try a base hydrolysis on the intimate mixture of RDX and HMX as a homogeneous reaction, which I did by dissolving the mixture in acetone and subjecting the resulting solution to base hydrolysis. It worked very well. The RDX was preferentially hydrolyzed, making it possible to set the conditions of hydrolysis to minimize the loss of HMX and maximize the hydrolysis of RDX. It was now possible to accurately determine the proportions of the two products produced by the Bachman process. This reaction could be carried out under a variety of conditions, and the most suitable production condition required to maximize the yield of RDX and minimize the amount of HMX was determined. This research turned out to be my PhD thesis, and it turned out to be a contribution to the war effort as well, while at the same time producing fundamental information about an important reaction. The papers dealing with this subject were classified during the war, but later they were published in 1952 in the *Canadian Journal of Chemistry*. I had optimistically persevered in my aim to become a scientist and was rewarded not only with a PhD, but also with a new experience, which served me well in my future scientific career.

The time I spent at McGill was my first experience working closely with a group of other scientists, 16 other young PhD students. It took a great deal of dedication, hard work, and esprit de corps to work successfully in the crowded laboratories available at that time at McGill University. Most students came from other provinces, because during that period McGill was only one of two Canadian universities that offered PhD degrees. We earned our keep as teaching assistants. We took graduate courses. We worked long hours into the night, including weekends. We helped each other and became close friends. Dr. Winkler was an excellent role model. He was even-tempered and friendly, and he worked very hard teaching and directing the research of his 17 graduate students.

Although I have not continued to pursue these types of kinetic studies of organic reactions during my post-PhD years, the work I did at McGill was

nevertheless very important as preparation for my career in the years that followed.

I received my PhD in 1944 and was immediately directed to join the University of Montreal Project. This was a combined European-Canadian effort in research on nuclear fission. The laboratory was under the direction of Professor Cockroft, and it included an extremely talented group of scientists from the European scientific community. I had just landed in a strange world and was about to be introduced to a new field, nuclear chemistry, a completely new experience for me.

I was assigned to become the third member of a team composed of Drs. W.J. Arrol and K.F. Chackett, whose job was to determine the concentrations of the noble gases argon, krypton, and xenon that are present in uranium metal irradiated with slow neutrons. Argon was introduced into the metal during its manufacture, whereas the isotopes of krypton and xenon were formed as products of uranium fission. Our job was to develop techniques to extract and measure the concentrations of the tiny amounts of these gases dissolved in the uranium metal. The purpose of these analyses was to determine whether the concentrations of these gases were large enough to introduce dangerous stresses in the uranium metal rods. Such stresses could cause an undesirable disintegration of the uranium rods in the reactors.

Bill Arrol was highly skilled and experienced in dealing with tiny amounts of noble gases, including helium. Ken Chackett was a recent PhD graduate. Some of the equipment needed for this type of work was not commercially available at the time. For example, we had to regrind our own glass stopcocks and build equipment such as mercury valves whenever they were needed. My first job was to acquire the needed skills to help build the vacuum extraction lines. Consequently, I spent a great deal of time in the first few months at the glass-blowing table learning how to do this. I became quite a respectable glass blower, a skill that serves me well to this very day. In time we succeeded in isolating very clean samples of a mixture of noble gases. We separated them into their pure components by taking advantage of the differences in their degrees of absorption on activated charcoal at low temperatures.

Working in the University of Montreal laboratory was somewhat more relaxing than teaching, taking courses, and doing research for my PhD at McGill had been, and a more reasonable social life was possible. It was about this time that I met my wife, Diane, on a blind date arranged by Diane's sister. It didn't take long for me to appreciate this lovely lady. In September of 1946 we got married.

Professor Harry Thode from McMaster University was one of the Canadians active in the Atomic Energy Project of Canada at that time. He suggested analyzing the mixture of the noble gases we extracted from the uranium sample in his mass spectrometer, the only one then in existence in all of Canada. His

rather simple mass spectrometer analyses allowed him to identify the abundances of all the isotopes represented by the masses of krypton and xenon that were produced by fission. In this way he was able to determine the fission yields of the atomic products produced by the fission of  $^{235}\text{U}$ . The isotopes of these noble gases were the final nonradioactive products produced from the initial radioactive fission products. This was a dramatic illustration of how a single mass spectrometer scan of a mixture of gases could accomplish what many radiochemists in both the United States and Canada were trying to determine by measuring the detailed decay schemes of the initial fission products.

I was particularly fortunate to become not only an admirer, but a life-long friend of Harry Thode. After the war, the University of Montreal Laboratory was about to be shut down and the Canadian Atomic Energy Program was going to move to the Chalk River site in Ontario. Harry Thode invited me to come to his laboratory, where we would continue to study the isotopic compositions of the noble-gas fission products as they were affected by varying conditions of neutron radiation. This type of experiment could provide information about some of the nuclear processes involved in fission, a topic of fundamental scientific interest.

Although I moved to McMaster University in 1946, I remained an employee of the Canadian Atomic Energy Project. At McMaster University, we proceeded to build the necessary extraction lines to carry out our project. However, our ability to do this work depended entirely on the cooperation of the American Atomic Energy Commission, which had previously provided the irradiated uranium. Unfortunately, because of the spy scare at that time, involving the defection of a Cypher Clerk in the Soviet Embassy in Ottawa, the American government was reluctant to ship such material into Canada. Because I was still an employee of the Canadian Atomic Energy Project, I was destined to move to the Chalk River facility. Diane and I visited Chalk River and saw some of the advantages of making that our permanent home, including beautiful quarters and a community of people with common interests. However, while I was still at McMaster University, Harry Thode received a phone call from Professor Harold Urey at the University of Chicago inquiring if he knew of a young PhD who would be willing to come to Chicago to work with him on a problem dealing with paleoclimates.

Harry Thode had worked with Harold Urey at Columbia University and had benefited very much from his experience, so he recommended me to Urey for this job. Harold Urey subsequently sent me a letter offering me a postdoctoral fellowship at a given salary, as well as living quarters in his garage apartment. The opportunity to work with Urey and spend time at the University of Chicago was intriguing. I accepted his offer, and after a time, and with the help of Dr. Thode, I got a one-year visa allowing me to go to Chicago.



The less said about Chicago, with respect to the living quarters available to a newly arrived postdoctoral fellow, the better. The personnel at the University of Chicago, however, were outstanding. Robert M. Hutchins, the president of the University of Chicago, had accumulated an outstanding faculty, who in turn had also attracted a very talented group of young postdoctoral fellows, all of whom were eager to do fundamental research, having spent so many years working on war related problems. Harold Urey himself was eager to do some new research on a problem that, as he put it, "was not useful." He chose to find out whether a marked temperature change had occurred on Earth 65 million years ago could have been responsible for the extinction of the dinosaurs. He wanted to do this by measuring the temperature of the ancient ocean by means of the isotopic records of oxygen in ancient marine fossils. He published this idea and estimated that, based on the existing accuracy of measuring oxygen isotopic compositions, he might be able to measure this temperature to an accuracy of 6°C (Urey 1947). This sort of problem apparently had not interested the bright physics and chemistry postdoctoral fellows already at Chicago, and thus Urey turned to Canada and got me. As a result of my interests and experience in geology as an undergraduate student I had the advantage of appreciating this problem. I was also willing to learn something new about stable isotope geochemistry.

Diane and I moved to Chicago in the fall of 1947. Our apartment above the Ureys' garage had many problems, but it was adequate considering how little was available for temporary visitors in an area within a reasonable distance from the University of Chicago. In addition, I had reliable transportation to work and back: Urey and I were chauffeured by Mrs. Urey in a car Urey had bought with the Nobel Prize money he had won. Of course, having a friendly close neighbor was also desirable to us: I was available when Urey wanted to test some of his ideas on someone, and Mary Urey was available as a baby-sitter after our son was born in 1948.

The scientific community at the University of Chicago at that time was composed of a remarkable mixture of renowned chemists, physicists, and even biophysicists. In addition to these major scientists, Chicago had recently appointed a number of brilliant young junior faculty whose contributions to science over the years to come were quite remarkable. A faculty of this type attracted a large group of excellent postdoctoral fellows and graduate students. A major fraction of this highly talented faculty belonged to the Institute for Nuclear Studies, and their labs and offices were all housed in a new building. Every Thursday afternoon, there were informal talks that consisted of important presentations of new ideas and information. The idea appeared to be to provide interactions and opinions from a variety of scientists on any topic of scientific interest. In addition, visiting scientists came from all over the world to present some of their

latest work. This was a wonderful experience for a young scientist, a place where a researcher could get a feeling of excitement and commitment while learning something new and topical. The subjects we discussed ranged from the fundamentals of nuclear physics and chemistry, to processes that governed the origin of the Earth and the Universe. The temperature of the ocean millions of years ago, the project I was working on, was included as one of the topics.

The principal collaborators on Harold Urey's paleotemperature project included Charles McKinney (an engineer who had supervised the construction of a duplicate of the remarkably precise and accurate mass spectrometer developed by Al Nier at the University of Minnesota), John McCrea (Urey's only graduate student), an electronic technician named Harold Allen, and myself. We realized that it was necessary to upgrade the electronics needed for the mass spectrometer. What was required was a stable high-voltage source for the acceleration of the ions, a power supply able to produce an appropriately stable magnetic field for the electromagnet, and a sensitive and stable electrometer for measuring the currents generated by the ion beams containing the different masses.

After some time, a number of false starts, and a lot of hard work, Urey's team was able to put together a functioning instrument with all of the above attributes. The mass spectrometer, on a good day, gave reasonable isotope measurements such that  $^{18}\text{O}/^{16}\text{O}$  ratios of  $\text{CO}_2$  gas could be measured relative to a given standard gas, to within  $\pm 0.20$  per mil (parts per thousand) (McKinney et al 1950). Some time later, we were able to obtain a vibrating reed electrometer that allowed us to make routine measurements to  $\pm 0.10$  per mil.

The important issue in all this work was that we were able to upgrade the Nier mass spectrometer enough to measure variations in  $\delta^{18}\text{O}$  by a factor of 6 more accurately than Harold Urey had stated would be necessary in his published paper on the topic. In principle, therefore, we ought to have been able to measure the temperature at which calcium carbonate precipitated to within  $\pm 0.5^\circ\text{C}$ . This fact entirely changed the objectives of our research with respect to the problems involved in extracting pure  $\text{CO}_2$  from biogenic calcareous marine animals. At the same time, the increased precision of our mass spectrometer definitely raised interest in the issue of measuring marine paleoclimates. Unfortunately, at that critical time, Diane and I and our very young American son, Reuben, all had to return to Canada to try get a new immigration visa. Urey wanted me to return to Chicago to continue working on the paleotemperature project. Of course, I had already realized from my one-year experience there that the University of Chicago was a very special place. So, I also was eager to return, in spite of the fact that I was informed that I still had a job available to me at Chalk River in Canada. A great deal of patience from Diane, and the gracious hospitality of her parents, allowed us to wait out our time in Montreal before we were able to get the appropriate visas to return to the University of Chicago. I later found

out that Urey's intervention had a great deal to do with my being able to get this visa. We returned to Chicago in the fall of 1948, and I resumed my research. A short time after that, John McCrea received his PhD and accepted a job in Canada, and Charles McKinney moved to another position at the University of Chicago.

There were certain obvious problems associated with our project right from the start. It was important to extract  $\text{CO}_2$  from the calcium carbonate of shell material without modifying the isotopic composition of the oxygen by any reagent utilized in this process, and without affecting the reproducibility of the isotopic analyses to more than  $\pm 0.1$  per mil of the ratio. In addition, both modern and preserved fossil shells were known to contain mixtures of oxygen and hydrogen in the form of carbonaceous material that could affect the isotopic composition of the  $\text{CO}_2$  extracted from the shells. Any method that was to be used to purify the calcium carbonate of the shells could not be allowed to alter the isotopic composition of the calcium carbonate. One of the major problems was the fact that we had no information about what the isotopic compositions of the different sources of  $\text{CO}_2$  should be, including the  $\text{CO}_2$  from the shell calcium carbonate. Ralph Buchsbaum, a University of Chicago biologist, set up controlled experiments growing calcium carbonate shells, and Heinz Lowenstam, Professor of Paleontology at the University of Chicago, served as a consultant paleontologist. Heinz turned out to be a very close friend with whom I interacted a great deal at the University of Chicago and who later decided to join the California Institute of Technology (Caltech) Division of Geological Sciences at the same time that I did. As it turned out, all the possible sources of error did indeed produce serious problems, including that of organic sources of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  forming during the process of purification of the powdered shell materials. After considerable effort, all of these very difficult problems were solved. However, I learned the important lesson that if anything can go wrong it will, and I acted accordingly. We were able to destroy the organic matter by pyrolysis in a fast stream of helium by using a temperature high enough to destroy the organic matter but low enough that the associated calcium carbonate was not affected. Another problem that surfaced was that the helium we used contained some oxygen that had to be removed prior to the helium contacting with our sample. Finally, we reacted the purified calcium carbonate with concentrated phosphoric acid to produce  $\text{CO}_2$ . We finally published a reliable temperature scale (Epstein et al 1953) and used it to obtain a series of reliable paleotemperatures resulting in several published papers, which came out between 1951 to 1954 (Urey et al 1951, Lowenstam & Epstein 1954).

This was the beginning of a major field, and since those early efforts, hundreds and thousands of carbonate analyses have been carried out all over the world

using basically the same  $\text{H}_3\text{PO}_4$  technique that we developed in the early 1950s. For example, Cesare Emiliani (1955) who was a student at Chicago at that time used this technique on foraminifera in deep sea cores to establish the now universally used chronology of glaciations during the Pleistocene.

I realized that I could not be a perpetual postdoctoral fellow. I was 32 years old, and it was now some 8 years after my PhD degree. At the same time, I had a duty to my family to move into a more permanent position. Therefore, I began what I thought would be a lengthy process of looking for a new position. There was no obvious indication that there might be a possible position in the University of Chicago Geology Department. Those who were most interested in isotope geochemistry were the younger faculty, some of whom were not in permanent positions themselves. At about that time, I bumped into Harrison Brown, a Professor of Chemistry at the University of Chicago, in one of the halls in the Institute of Nuclear Studies and casually asked him if he was aware of any positions that might be available for the likes of me. Although this was probably one of the few times he and I had spoken during my entire time at Chicago, he replied, "Would you like to come to Caltech with me?" To some degree this reminded me of my coincidental contact with Alan Campbell at the University of Manitoba, when it had been decided that I would pursue a Master of Science degree with him. The present coincidence was even more surprising, since I wasn't quite ready to leave the University of Chicago at that time. I didn't quite agree to accept the offer, but I was interested enough to accept Harrison's invitation to come to Caltech to give a seminar.

I recall that it was on a very cold day in February in Chicago when I joined Clair Patterson and Charles McKinney on a train trip to Pasadena. In Pasadena, I was greeted by a beautiful, warm February day and by an interested faculty and student body who responded very enthusiastically to my seminar. Although my seminar dealt with the paleotemperature work, the topics of the isotopic compositions in precipitation and in ice deposits were also brought up. In particular, I spent some time after the seminar at the blackboard with Robert P. Sharp, Professor of Geology at Caltech, discussing various applications of stable isotopes to problems in climatology. The warm reception they gave and their enthusiasm toward what I might be able to contribute to this interesting and unusual geology department were very welcome.

When I returned to Chicago and informed Harold Urey that I had decided to take the job at Caltech and move to Pasadena, he offered to raise my salary if I would stay. I was very tempted to remain in Chicago for another year to take advantage of the many research projects that interested me using the new mass spectrometer and functioning laboratory facilities I had helped to develop there. However, good sense told me that I must make the change. In doing so, I probably lost a whole year of research time and the authorship of several

papers. However, I could not ignore such an obviously important opportunity that might not be available in the future.

In this brief report, I cannot possibly describe adequately the importance of my four-year residence at University of Chicago, as far as my preparation for a future in science was concerned. There were important science projects being done, and both the young and the old researchers seemed to take special pleasure in their work. They were stimulated by the scholarly environment that existed there. Harold Urey continuously came up with new insights or special calculations with regard to the origin of the Earth and the Solar System. At that time, he was in the midst of preparing to write a book about the origin of the planets. He liked to test his ideas on whomever was around at the moment, even if he didn't get the desired feedback. Of course, those of us who were there at that time learned a thing or two from our interaction with Urey.

As for the rest of the University of Chicago personnel, there were many young bright people, including graduate students, postdoctoral fellows, and young faculty pursuing research projects, whose subsequent impact on the science was monumental. Julian Goldsmith has written a prefatory article in this same series entitled "Some Chicago Georecollections" (Goldsmith 1991) and I won't repeat all that material here. However, Julian named some 50 or more young scientists, the majority of whom I knew and benefited from by interacting with them. It was a remarkable period at the University of Chicago and I was delighted to have been there during that time. My colleagues, the students, postdoctoral fellows, and the faculty at Chicago, were truly inspirational. It would be impractical to try to list everyone, but I wish to stress how important they all were to my future scientific career.

I do need to make a special comment about Harold Urey, whose patience and continuous support was so instrumental in my maturation as a scientist and as a scholar. There is no doubt that the success I attained after leaving Chicago was due in large measure to my interaction with him as his postdoctoral fellow. His broad interests and good taste in the choice of problems, and the dedication with which he pursued every avenue in solving these problems, were only some of the lessons I learned from him. I continued my contact with Urey when he and his family later moved to La Jolla, California. As it turned out, many of my friends also soon left Chicago, a significant fraction of them taking positions in the University of California system. In large measure as a result of these close ties, the West Coast geochemistry community organized yearly meetings at one of four different locations: the Universities of California at San Diego, Berkeley, and Los Angeles, and at Caltech. These meetings lasted for one day, allowing us to share mutual interests and to report progress in our research.

At Caltech, the Division of Geological Sciences was obviously making a serious move to set up new activities in the Earth sciences. This included a major concentration in geochemistry. Although my initial offer was as a research fellow, somehow I felt that I would be able to flourish at this private school. Although Harrison Brown was responsible for my coming to Caltech, and incidentally my initial salary was paid from his Atomic Energy Commission contract, he never treated me as his postdoctoral fellow, but rather as a colleague. After being a student for so many years, I finally had the opportunity to be a teacher and design research projects, which gave me the chance to develop my own style based on my past experiences. Within two years, I was promoted from research fellow to associate professor with tenure. I obviously had come to an ideal situation that specifically fit my inclinations and talents. My promotion to associate professor at Caltech was acknowledged by a very nice letter of congratulation from Harold Urey.

It is interesting to understand something about the consequence of Caltech's policy of introducing new activities into the geology division at this time. Prior to the arrival of Harrison Brown in 1951, the Division of Geological Sciences at Caltech consisted of 7 professors, 5 associate professors, and 1 assistant professor, a total of 13 faculty. Of these faculty, 3 ended up as members of the National Academy of Sciences. By 1956, 4 of these faculty had either retired or gone elsewhere. New appointments had increased the faculty to a total of 18, including 12 professors, 1 associate professor, 4 assistant professors, and 1 special senior research fellow. Of these 18 faculty members, 13 were eventually elected to the National Academy. Interestingly, these included the 4 newly appointed assistant professors during that period of time, namely Clarence Allen, Leon Silver, Gerald Wasserburg, and Barclay Kamb. The other new more-senior faculty members who joined Caltech during those five years, including Harrison Brown, Heinz Lowenstam, Frank Press, Clair Patterson, and myself, were also eventually elected to the National Academy. Much of the change that took place in the division during these years was a result of the vision and attitude of President Lee Dubridge, and of other senior faculty in our division under the leadership of Bob Sharp, who was chairman at the time these major changes were initiated. Bob Sharp had a special ability to look ahead and recognize what kind of scientific activity would be productive in the future. Caltech's philosophy at that time was discussed in a *Time Magazine* article in 1955 featuring Dr. Dubridge on its cover. The additions to what was already a distinguished faculty in the Geology Division were important to all of us and enabled us to apply our expertise and interests to many different kinds of geology. This dynamic and energetic faculty also supported major changes in our division over the years, such as the establishment of a major new research direction in planetary science in the 1960s.

Turning to my own work, I could clearly see that there were many natural processes that affect the disposition of our environment and that could be studied by taking note of their effect on the isotopic compositions of the lighter elements associated with these processes. A paper published by Toshiko Mayeda and myself (Epstein & Mayeda 1953) about some work done in Chicago pointed out an obvious consequence of the differences in the vapor pressure of the different isotopic species of water. Namely, as one observes in a regular multistage distillation column, the isotopic composition of the precipitation resulting from the condensation of the water vapor in the Earth's meteorological cycle is related to climatic temperature.

Once Caltech's mass spectrometer was built and was available for isotope analyses, we proceeded to analyze the  $\delta^{18}\text{O}$  of the waters originating from all the storms that passed over Pasadena for one year, from 1953 to 1954. These samples were collected, with the kind assistance of my wife, Diane, from a spout that allowed us to sample the water runoff from the roof of our house. The objective was to determine if the pattern of the isotopic records followed the expected relationship between the isotopic composition of the precipitation and the temperature changes associated with cold fronts, warm fronts, and occluded storms. As expected, for the cold front storm the  $\delta\text{D}$  decreased with time as the heavier cold air penetrated the area to a greater extent. It uplifted the warm moist air, causing adiabatic cooling and increasing the intensity and quantity of the rainfall. The reverse isotopic pattern was observed for the rain originating from the warm-front storm. The occluded storm had a minimum  $\delta\text{D}$  value as it passed over Pasadena. In some cases, there were unusual, rapid, and dramatic changes in  $\delta\text{D}$ , which had to be due to stratification of the cloud structures, with the low deuterium waters coming from clouds at a higher elevation. We also analyzed the  $\delta^{18}\text{O}$  records of rain collected in five different locations that were affected by a single storm system. These locations included Santa Barbara, Pasadena, and Mount Wilson. The isotopic records corresponded to their expected relationships with climatic temperatures in these areas (Epstein 1956).

We continued analyzing precipitation from other localities, including those in glacial deposits, which we did with the cooperation of Bob Sharp (Epstein & Sharp 1959). Our initial samples were obtained from both Greenland and from Antarctica and were taken from the walls of trenches. Their isotopic compositions showed strong seasonal variations. Again, the simple difference in the vapor pressure of the different isotopic species of water provided valuable information regarding rates of snow fall and climatic temperatures of the past. This work was continued by glaciologists in Greenland and in the Antarctic. Their skill and experience were of paramount importance to do the necessary research in this field. Although this was obviously very important research that I might have continued, several other considerations came into play. Namely,

there were other processes occurring in nature that were just as interesting and important and probably more compatible with my experience, as well as with the needs of the students and postdoctoral fellows at Caltech. Consequently, one of my major interests was to expand the new and exciting approach of stable isotope research to many natural processes that occur in both terrestrial and extraterrestrial situations.

Caltech turned out to be an ideal place to achieve these objectives. For example, Bob Clayton was a newly arrived graduate student in the Division of Chemistry and Chemical Engineering at that time. He found a project working with me that suited his taste and interests, and he decided to do his PhD thesis under my direction. The project we decided to do was to determine how the  $^{18}\text{O}/^{16}\text{O}$  ratios were partitioned among the different minerals in igneous and metamorphic rocks, with the idea that this might reveal something about the temperature conditions under which the rocks were formed. Estimating the temperatures at which the rocks and minerals were formed was a major interest at that time in the geology community. In addition, Bob Clayton wanted to try a different technique of extracting the oxygen from the minerals for isotope analyses. This involved high-temperature reduction of the silicates with powdered graphite in order to complement and check the validity of the extraction technique with which the silicates were reacted with fluorine, a very dangerous and corrosive gas (Clayton & Epstein 1958). The fact that Clayton was a very talented student also gave me the opportunity to learn a few things from him. Bob Clayton is now a distinguished professor at the University of Chicago and has received many awards for his wonderful discoveries. Beyond the pleasure that I always get from the success of my students, I feel that Bob's success at the University of Chicago is particularly satisfying, considering my past affiliation with that university.

During my early days at Caltech, an adventurous biology student named Rod Park decided to take my course in geochemistry and to do part of his PhD thesis research with me. His decision to work with me was encouraged by his advisor, Jim Bonner. After some discussion, we decided to determine the mechanism for the  $^{13}\text{C}$  fractionation that occurs during  $\text{CO}_2$  fixation by plants (Park & Epstein 1960). We published two papers on this subject, and my fascination with this type of biogeochemistry has stayed with me over the years. Another biology student, Michael DeNiro, decided to do research with me some 15 years later, and his work also resulted in a PhD degree from the Division of Biology, as well as our joint publication of several papers (e.g. DeNiro & Epstein 1978). Both of these scientists were appointed to the faculties of the University of California system and have earned distinguished careers.

Hugh Taylor was an undergraduate student who became the first geochemistry major at Caltech. He did some laboratory work with me and wrote a



term paper for one of my courses. Hugh originally left to do graduate work at Harvard but decided he was more interested in getting involved in stable isotope research, so he returned to Caltech to do his PhD with me. He earned his PhD degree with flying colors. The list of papers we eventually published together is too long to include here. After his thesis work was done, it became obvious that he was an unusually talented research scientist and teacher, so after again leaving Caltech for a short period of time to go to Penn State, he was invited to come back to join our faculty. Hugh Taylor was one of the most important appointments we have made. During the 1960s our joint research projects produced some 12 papers dealing with a variety of rocks, including meteorites and tektites (Taylor & Epstein 1962a,b, Taylor et al 1965).

The 1960s was a very productive period overall. In addition to the joint projects shared with Hugh Taylor, my own research involved applying isotopic records to a variety of problems dealing with processes under which different types of rocks and minerals were formed. Three postdoctoral fellows were involved in solving problems relevant to the origins of carbonates. They were James O'Neil (O'Neil & Epstein 1966), presently at the University of Michigan, and Egon Degens and Joachim Hoefs, who were visitors from Germany. We also proceeded to investigate the systematics of the distribution of the  $^{18}\text{O}/^{16}\text{O}$  ratios in metamorphic rocks and of rocks that were involved in hydrothermal mineralization. Don Garlick undertook this project as his PhD thesis, producing several important papers on the subject (Garlick & Epstein 1967).

Sam Savin, currently a dean at the Case Western Reserve University, did his PhD thesis on the isotope geochemistry of oxygen and hydrogen in clays and ocean sediments (Savin & Epstein 1970). Bruce Smith, a biology postdoctoral fellow, started working with me on the stable isotopes of hydrogen and carbon in a salt marsh biota as well as on measuring the carbon isotopic ratio distribution among the higher plants (Smith & Epstein 1970).

Stable isotope geochemistry was obviously making some inroads with a variety of students from different disciplines. All of them realized the versatility inherent in this approach, and the marked differences in objectives that can be pursued with stable isotopic data.

It was at this time that lunar samples became available for analysis. Hugh Taylor and I were invited to participate in the program dealing with the stable isotope geochemistry of these samples. This was a wonderful opportunity to participate in a new and exciting research program that caught the imagination of the whole world. It was also obvious that the techniques we developed over the years could provide very useful information about the unique origin and history of the lunar samples. This was a very productive stage for both Hugh Taylor and myself. One of the highlights that our research produced was to show the almost complete absence of deuterium in hydrogen gas extracted

from the lunar soils (Epstein & Taylor 1970a). This indicated that essentially all the hydrogen on the Moon was of solar origin and had been implanted by the solar wind. This result also showed that the Sun preferentially burned all the deuterium in its nuclear furnace. In addition, we found that the surface of the Moon has been bombarded with particles that preferentially vaporized both  $^{32}\text{Si}$  and  $^{16}\text{O}$  from the silicate grain surfaces of the lunar soil, leaving a residue that was tremendously enriched in the heavy isotopes of both elements. This also resulted in a higher Si/O ratio in the surface of the lunar grains. Once this very heavy surface oxygen and silicon were preferentially removed by partial fluorination of the lunar soil, the isotopic composition of the body of the soil and the rocks on the Moon were found to be identical to that of the Earth (Epstein & Taylor 1970a,b, Epstein & Taylor 1971).

In the 1970s we started a project dealing with the isotopic composition of hydrogen and oxygen in silica from the Deep-Sea Drilling Project as well as from bedded ancient cherts. This was undertaken by Paul Knauth, who is at present a professor at Arizona State University. The combination of measuring the isotopic compositions of oxygen and hydrogen from the same mineral provided temperature information about the origin of these cherts going back to Precambrian time (Knauth & Epstein 1976). Paul Knauth continued and expanded upon these studies of Precambrian cherts. Similar Precambrian temperature information was later obtained from oxygen isotope records extracted from coexisting cherts and phosphates (Karhu & Epstein 1986).

During the late 1970s, we initiated two types of research projects that we are continuing to this day. One involved the extraction of climatic temperature data from the isotopic composition of unexchangeable hydrogen in cellulose extracted from trees. We have shown that the isotopic composition of hydrogen extracted from total wood introduced complications as a consequence of the fact that different compounds of wood are isotopically highly variable. Consequently, our laboratory procedure involves isolating cellulose that has been stripped of the isotopically exchangeable hydrogen to form nitrocellulose. With that in place, we were able to obtain reasonably good climatic records from the isotopic compositions of the hydrogen in trees. A series of papers, started in 1976 with Crayton Yapp, showed that there is a good relationship between the isotopic composition of hydrogen in nitrocellulose and mean annual temperature (Yapp & Epstein 1982). This work was continued through the 1980s with Crayton Yapp, R.V. Krishnamurthy, and myself, and in the 1990s with a series of papers published with Xiahong Feng (Feng & Epstein 1994, 1995). These three scientists now have faculty positions at Southern Methodist University, at the University of Michigan, and at Dartmouth College, respectively. In these papers, we have analyzed consecutive tree rings for the purpose of comparing their  $\delta\text{D}$  values with the historically recorded temperatures in the same area.

We have also published a paper on isotopic records for 14 trees covering a latitudinal and temperature range between Kenya and Alaska. Our findings suggest that warming had taken place in the nineteenth century, whereas somewhat more erratic climatic records were observed for the twentieth century (Feng & Epstein 1996). In this connection, we have been able to obtain information from fragments of wood that are thousands of years old. These fragments could not be used for tree ring chronology, but they provided climatic information for the times designated by their C-14 dates. An interesting result was obtained from the isotopic composition of hydrogen extracted from C-14 dated fragments of willow trees that grew on the North Slope of Alaska (Epstein 1995). These isotopic records match those of the ice cap on Greenland, confirming the ability of a fragment of wood to record the isotopic composition of the local precipitation. The consequence of these data, in terms of the climatic significance of the isotopic records in the Younger Dryas, contradicts the normal interpretation of the isotopic records of the ice cap on Greenland. These research projects are still ongoing and have significance in evaluating past global changes.

The other major activity at our laboratory that was started in the late 1970s dealt with the isotopic composition of hydrogen in the organic matter of meteorites. For example, Francois Robert (Robert & Epstein 1982), Richard Becker (Becker & Epstein 1982), and I reported some results on hydrogen, carbon, and nitrogen isotopes in the organic material extracted from meteorites, and Richard Becker and I analyzed solvent extractions from carbonaceous chondrites. However, the main results originated from several research activities dealing with the concentration and isotopic composition of organic matter isolated from HF-HCl residues of carbonaceous meteorites. This work was done with Dr. J. Yang (Yang & Epstein 1983, 1984), a graduate from the University of Chicago. We discovered a remarkably large variation in the isotopic composition of hydrogen and carbon in residues that have been stepwise oxidized. For example, the initial HF-HCl residue was subjected to  $H_2O_2$  oxidation, which preferentially resulted in the destruction of organic fractions having a low D/H ratio. In addition, highly  $^{13}C$ -enriched silicon carbide grains were also discovered in this residue.

One of our projects was initiated by a coincidental meeting with Dr. John Cronin, professor of Chemistry at Arizona State University. I attended a lecture that he was giving in our Division of Biology, and listened with great interest to the description of his work on amino acids in meteorites. In earlier days, I had had the impression that there was a controversy about the origin of the amino acids and that they were actually of terrestrial origin. I approached Dr. Cronin about the possibility of obtaining tiny amounts of his amino acid samples for analysis. He sent us the samples and we were able to analyze this material, finding that the D/H ratios in these samples were at least twofold-enriched compared with terrestrial values. This was certainly very strong evidence that the amino acids extracted from the Murchison meteorite had been formed,

at least in part, in interstellar space (Epstein et al 1987). Further cooperation between our two laboratories resulted in five interesting papers dealing with the amino acids, hydrocarbons, monocarboxylic acids, ammonia, amines, and polar hydrocarbons extracted from a clean central part of the Murchison meteorite (Cronin et al 1993, Krishnamurthy et al 1992, Pizzarello et al 1994).

Another project we initiated grew out of providing a part-time job for the wife of a visiting scientist. An interesting experiment resulted, dealing with the effect of respiration on the isotopic composition of oxygen. We know that oxygen in the atmosphere is enriched in  $^{18}\text{O}$  as a consequence of the preferential utilization of the  $^{16}\text{O}$  by the Earth's biota. We tried to determine whether humans contribute to this enrichment. The experiment simply compared the isotopic composition of atmospheric  $\text{O}_2$  with the isotopic composition of  $\text{O}_2$  exhaled by humans. Indeed, we found that humans enrich  $^{18}\text{O}$  by their respiration. The degree of enrichment depends on the fraction of inhaled  $\text{O}_2$  they utilized, the condition of their lungs as a result of smoking, and the intensity of their physical exercise. The fraction of the  $\text{O}_2$  that they used is measured by comparing the  $\text{O}_2/\text{N}_2$  ratios in their breath samples with that ratio in the atmosphere (Epstein & Zeiri 1988).

Laurie Leshin (Watson), a graduate student with Ed Stolper and myself, examined the isotopic compositions of hydrogen and carbon from the SNC (shergotite, nakhlite, chassignite) meteorites that have been proposed to have originated from Mars. Analyses of the water in micas, with the cooperation of Dr. Hutcheon in Dr. Wasserburg's laboratory, gave a D/H ratio enriched by fivefold compared to terrestrial ratios (Watson et al 1994). This effect was observed to a lesser degree in the water extracted from whole-rock SNC meteorite samples. These results provide confirming evidence for a Martian origin of the SNC meteorites (Leshin et al 1996).

My cooperative studies with Ed Stolper have also included projects involving the determination of the isotopic fractionation of oxygen between minerals, using an indirect technique in which the two minerals are exchanged with a small amount of  $\text{CO}_2$  at the desired temperature (Matthews et al 1994). We have also done some work on the solubility of water in glass and the isotopic fractionation associated with the degassing of hot volcanic glasses (Newman et al 1988).

Another colleague, Tom Ahrens, studies the shock-induced devolatilization of a variety of materials to determine the effect of impact by meteorites on the Earth's surface. We have studied the isotopic compositions of the  $\text{CO}_2$  produced by shocked calcium carbonate (Boslough et al 1982) and the  $\text{H}_2\text{O}$  produced by shocked serpentine (Tyburczy et al 1990). The isotopic analyses of the products of these shock-induced devolatilizations indicate that these events generally cause isotopic fractionation. Hopefully, the relationship between the isotopic compositions of the volatiles and that of the starting material may provide some information about the nature of the original material that survives the impact from an incoming meteorite.

It should be obvious to anyone who reads this sketch of my career that I was fortunate to have crossed the paths of so many great and generous scientists. In their presence I have learned much and their influence has affected my whole career. The experience of having to deal with projects that required unusual and special innovative approaches was of great benefit in solving problems and added a special flavor to my scientific life. When I came to Caltech I was ready to carry out a variety of research projects. I was also prepared to better communicate with colleagues and contribute to the different types of research that were carried out both in and outside of our division. Much of my success at Caltech was due to the quality of our students, but even more importantly it was due to the high quality of my colleagues in our division and in Caltech as a whole. It was in this milieu that ideas were created and nurtured until they flowered, producing research ideas worth pursuing to their fruition. It is with thanks to my colleagues that I end this biographical sketch.

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